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Valence-band Offsets of III–V Alloy Heterojunctions

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Valence band offsets at the heterointerface of lattice-matched alloy semiconductors are investigated with theoretical calculation, which is based on average bond energy theory in conjunction with a cluster expansion method. The predicted relative valence band positions of a wide range of III–V alloys are presented. The variation law of valence band offsets with composition is studied. Some trends of relative valence band positions are also presented. The theoretical results are in very good agreement with relevant experimental data. The table and figures summarizing the variation of valence band positions should be very useful in the design of novel heterostructure electronic and optical devices.

KEYWORDS: semiconductor; alloy; valence band offset; heterointerface; average bond energy theory; self-consistent calculation

INTRODUCTION

Band offsets at the heterointerface of two semiconductors, especially for heterojunctions involving lattice-matched alloys that are widely used in optoelectronic devices, are very important in device design and performance analyses.[1,2] Unfortunately, new device designs often require this information for alloy combinations for which no experimental data are available. Consequently, device designers are forced to perform several trial-and-error experiments. So, the theoretical prediction of band offsets at the heterinterface of two alloy semiconductors can provide directions for related experiments and device design.[3,4]

In this paper, we have systematically calculated the valence band offsets between most III–V semiconductor alloys via a self-consistent band structure method. Chemical trends in the determination of valence band offsets are discussed.

COMPUTIONAL METHOD

We have used the LMTO-ASA (linear muffin-tin orbital with atomic-sphere approximation) method to calculate the band structures of ordered ternary alloy $A_lB_{4-l}C_4$. Among the five ordered structures ($l = 0, 1, 2, 3, 4$), $l = 0$ and 4 are zinc blende (ZB) structures, $l = 2$ is CuAu (labelled $L1_0$) and $l = 1$ and 3 are luzonite ($L1_2$) structures.[5] The lattice constants of the five ordered structures can be obtained as the average of the bulk materials A, B and C in proportion to their contents, i.e. according to the well-known Vegard's law:[6] $a_{A_lB_{4-l}C} = (1 - l/4)a_{AB} + (l/4)a_{AC}$. On treatment of the cation's shallow d-orbitals for III–V semiconductors, we take the outermost d-electrons of the cations (such as Ga 3d, In 4d electrons and d-electrons of Zn) as valence electrons and the d-electrons of anions as core electrons, respectively,[7] for the band structure calculations of the five ordered structures. The special K-point method[8] is adopted for summation over the Brillouin zone.

After getting the self-consistent calculation, we calculate the average bond energy $E_m(x)$ according to the average bond energy theory (ABE) presented by Wang *et al.*[2,7]

$$E_m = (E_a + E_b)/2 \tag{1}$$

Where the bonding orbital energy E_b and antibonding orbital E_a can be expressed as

$$E_b = \frac{1}{4N}\sum_{n=1}^{M}\sum_{k} E_n(k) \tag{2}$$

$$E_a = \frac{1}{4N}\sum_{n=M+1}^{2M}\sum_{k} E_n(k) \tag{3}$$

Here $E_n(k)$ is the eigenvalue of n band at k point, N is the number of unit cells and M is the number of valence bands. For the ZB, $L1_0$ and $L1_2$ structures, M is evaluated by 4, 8 and 16, respectively. By using the average bond energy $E_m(x)$ as an energy reference, we can obtained the relative valence band position $E_{vm}(x)$ (so-called 'valence band offsets parameter') of disordered alloy systems as $E_{vm}(x) = E_v(x) - E_m(x)$, where $E_v(x)$ is the valence band maximum. By aligning the average bond energy in alloys I and II, the valence band offsets (VBO) parameter at a heterojunction such as alloy I/alloy II can be obtained as

$$\Delta E_v(x) = E^{II}_{vm}(x) - E^{I}_{vm}(x) \tag{4}$$

Using this equation, one can conveniently obtain the band offsets between the lattice-matched alloys with the

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Table 1. Results of valence band maximum $E_v(x)$, average bond energy $E_m(x)$ and VBO parameter $E_{vm}(x)$ for ternary alloys

Alloys	Lattice constant	$E_v(x)$	$E_m(x)$	$E_{vm}(x)$
$Ga_xAl_{1-x}P$	$5.467-0.020x$	$-0.036x^2+0.208x-1.123$	$-0.058x^2-0.335x-0.178$	$0.023x^2+0.542x-0.945$
$Ga_xAl_{1-x}As$	$5.660-0.006x$	$-0.100x^2+0.230x-1.180$	$-0.102x^2-0.298x-0.665$	$0.002x^2+0.528x-0.515$
$Ga_xAl_{1-x}Sb$	$6.136-0.041x$	$-0.008x^2+0.262x-1.245$	$-0.003x^2-0.188x-1.158$	$-0.005x^2+0.449x-0.087$
$In_xAl_{1-x}P$	$5.467+0.402x$	$-0.138x^2-0.271x-1.141$	$-0.036x^2-0.861x-0.189$	$-0.103x^2+0.590x-0.952$
$In_xAl_{1-x}As$	$5.660+0.419x$	$-0.085x^2-0.356x-1.182$	$-0.011x^2-0.834x-0.667$	$-0.073x^2+0.479x-0.515$
$In_xAl_{1-x}Sb$	$6.136+0.343x$	$-0.006x^2-0.396x-1.245$	$0.018x^2-0.661x-1.158$	$-0.024x^2+0.266x-0.087$
$In_xGa_{1-x}P$	$5.447+0.422x$	$-0.108x^2-0.497x-0.945$	$-0.055x^2-0.466x-0.565$	$-0.053x^2-0.031x-0.380$
$In_xGa_{1-x}As$	$5.654+0.404x$	$-0.079x^2-0.497x-1.046$	$-0.043x^2-0.409x-1.062$	$-0.036x^2-0.089x+0.016$
$In_xGa_{1-x}Sb$	$6.095+0.384x$	$-0.034x^2-0.624x-0.990$	$-0.005x^2-0.448x-1.348$	$-0.029x^2-0.175x+0.358$
$AlAs_xP_{1-x}$	$5.467+0.193x$	$-0.154x^2+0.128x-1.147$	$-0.138x^2-0.323x-0.197$	$-0.015x^2+0.451x-0.950$
$AlSb_xP_{1-x}$	$5.467+0.669x$	$-0.319x^2+0.204x-1.136$	$0.027x^2-0.997x-0.189$	$-0.346x^2+1.201x-0.947$
$AlSb_xAs_{1-x}$	$5.660+0.497x$	$-0.322x^2+0.240x-1.171$	$-0.151x^2-0.357x-0.658$	$-0.172x^2+0.597x-0.513$
$GaAs_xP_{1-x}$	$5.447+0.207x$	$-0.012x^2-0.089x-0.946$	$0.024x^2-0.522x-0.565$	$-0.036x^2+0.434x-0.381$
$GaSb_xP_{1-x}$	$5.447+0.648x$	$-0.278x^2+0.223x-0.943$	$0.016x^2-0.800x-0.565$	$-0.293x^2+1.023x-0.378$
$GaSb_xAs_{1-x}$	$5.654+0.441x$	$-0.285x^2+0.382x-1.062$	$-0.108x^2-0.141x-1.076$	$-0.177x^2+0.523x+0.014$
$InAs_xP_{1-x}$	$5.869+0.189x$	$0.017x^2-0.090x-1.548$	$-0.428x-1.084$	$0.017x^2+0.338x-0.464$
$InSb_xP_{1-x}$	$5.869+0.610x$	$-0.135x^2+0.032x-1.547$	$0.042x^2-0.759x-1.084$	$-0.177x^2+0.792x-0.463$
$InSb_xAs_{1-x}$	$6.058+0.421x$	$-0.093x^2+0.067x-1.621$	$-0.006x^2-0.283x-1.512$	$-0.086x^2+0.350x-0.109$

results of the relative VBO parameter from first-principle calculation. To calculate the VBO parameter $E_{vm}(x)$ of disordered alloy systems, the cluster expansion method is needed. In terms of the data of five ordered structures of ternary alloy $A_lB_{4-l}C_4$ and using the cluster expansion method, the valence band maximum $E_v(x)$ and average bond energy $E_m(x)$ of disordered alloy systems can be obtained by: $E_v(x)=\Sigma_l P_l(x)E_v^l$ and $E_m(x)=\Sigma_l P_l(x)E_m^l$ (here, l is a superscript and not an exponent). Where the statistical weight $P_l(x)$ is the probability that the l short-range ordered structure occurs in the alloy, it can be expressed as: $P_l(x)=\binom{4}{l}x^l(1-x)^{4-l}$.

RESULTS AND DISCUSSIONS

Band offsets parameter of alloys

The results of $E_v(x)$ and $E_m(x)$ for disordered alloy systems are given in Table 1. From this table one can find that the average bond energy $E_m(x)$ of III–V alloys is a function of the composition x, and it decreases almost linearly with x for most III–V alloys presented here. The results of $E_{vm}(x)$ for disordered alloy systems are also given in Table 1.

Most $E_{vm}(x)$ curves of III–V alloys are nearly linear, i.e. the bending of the $E_{vm}(x)$ curves is very small (<0.1), but that of several alloys such as $In_xAl_{1-x}P$, $AlSb_xP_{1-x}$, $AlSb_xAs_{1-x}$, $GaSb_xP_{1-x}$, $GaSb_xAs_{1-x}$ and $InSb_xP_{1-x}$ is a little larger (>0.1 but <0.4). In general, the VBO parameter $E_{vm}(x)$ of III–V alloys shows nearly linear relations abiding Vegard's law.[6] They are very different from that of $(III-V)_x(IV_2)_{1-x}$ systems such as $(GaAs)_x(Ge_2)_{1-x}$ and $(AlAs)_x(Ge_2)_{1-x}$ alloys,[4] for which the VBO parameter $E_{vm}(x)$ shows anomalous non-linear behaviour.

Valence band offsets between III–V alloys

Band offsets of $Ga_xAl_{1-x}M/Ga_yAl_{1-y}M$ (M = P, As, Sb) systems. The heterojunction $Ga_xAl_{1-x}M/Ga_yAl_{1-y}M$ (M = P, As, Sb) systems are very important III–V systems. Their band offsets have been studied widely by experimental method[17,18,20,21] and theoretical calculation[16] recently. The band offsets of these systems are listed in Table 2. The variations in the VBO at $Ga_xAl_{1-x}M/Ga_yAl_{1-y}M$ (M = P, As, Sb) systems with composition are shown in Fig. 1. From Fig. 1 and Table 2, one can see that the band offsets of $Ga_xAl_{1-x}M/GaM$ (M = P, As, Sb) systems are nearly linear. All of them decrease with the composition x when anions are changed from P to Sb, i.e.

$$\Delta E_v^{Ga_xAl_{1-x}Sb/GaSb}(x) < \Delta E_v^{Ga_xAl_{1-x}As/GaAs}(x) < \Delta E_v^{Ga_xAl_{1-x}P/GaP}(x) \quad (5)$$

Band offsets of $In_xAl_{1-x}M/In_xGa_{1-x}M$ (M = P, As, Sb) systems. Another kind of important lattice-matched heterojunction system is $In_xAl_{1-x}M/In_xGa_{1-x}M$ (M = P, As, Sb). The band offsets of lattice-matched heterojunctions $In_xAl_{1-x}P/In_xGa_{1-x}P$, $In_xAl_{1-x}-As/In_xGa_{1-x}As$ and $In_xAl_{1-x}Sb/In_xGa_{1-x}Sb$ are presented in Table 3 and Fig. 2. We can see that the band offsets of $In_xAl_{1-x}M/In_xGa_{1-x}M$ (M = P, As, Sb) systems are nearly linear, and they decrease with composition x. Similar to $Ga_xAl_{1-x}M/GaM$ (M = P, As, Sb) systems, the band offsets of $In_xAl_{1-x}M/In_xGa_{1-x}M$ systems decrease when the anions changed from P to Sb. It can be written as

$$\Delta E_v^{In_xAl_{1-x}Sb/In_xGa_{1-x}Sb}(x) < \Delta E_v^{In_xAl_{1-x}As/In_xGa_{1-x}As}(x) < \Delta E_v^{In_xAl_{1-x}P/In_xGa_{1-x}P}(x) \quad (6)$$

Band offsets of other lattice-matched systems. There are several III–V lattice-matched alloy heterojunctions

Table 2. Theoretical results of VBO for lattice-matched ternary alloy heterojunction $Al_xGa_{1-x}M/Al_yGa_{1-y}M$ (M = P, As, Sb) systems

Heterojunction	$\Delta E_v(x,y)$
$Ga_xAl_{1-x}P/Ga_yAl_{1-y}P$	$0.023y^2+0.542y-0.023x^2-0.542x$
$Ga_xAl_{1-x}As/Ga_yAl_{1-y}As$	$0.002y^2+0.528y-0.002x^2-0.528x$
$Ga_xAl_{1-x}Sb/Ga_yAl_{1-y}Sb$	$-0.005y^2+0.449y+0.005x^2-0.449x$

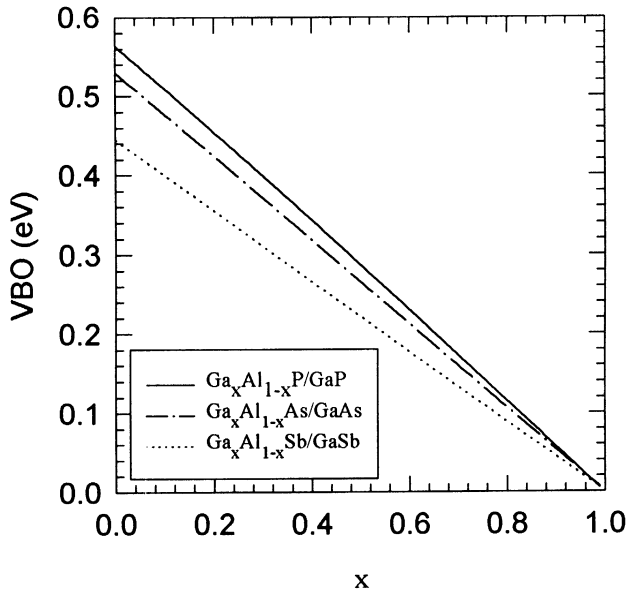

Figure 1. Variation in the valence band offsets (VBO) at $Ga_xAl_{1-x}M/Ga_yAl_{1-y}M$ (M = P, As, Sb) systems with composition.

Table 3. Theoretical results of VBO for lattice-matched ternary alloy heterojunction $In_xAl_{1-x}M/In_xGa_{1-x}M$ (M = P, As, Sb) systems

Heterojunction	$\Delta E_v(x)$
$In_xAl_{1-x}P/In_xGa_{1-x}P$	$0.050x^2 - 0.621x + 0.572$
$In_xAl_{1-x}As/In_xGa_{1-x}As$	$0.037x^2 - 0.567x + 0.531$
$In_xAl_{1-x}Sb/In_xGa_{1-x}Sb$	$-0.005x^2 - 0.441x + 0.445$

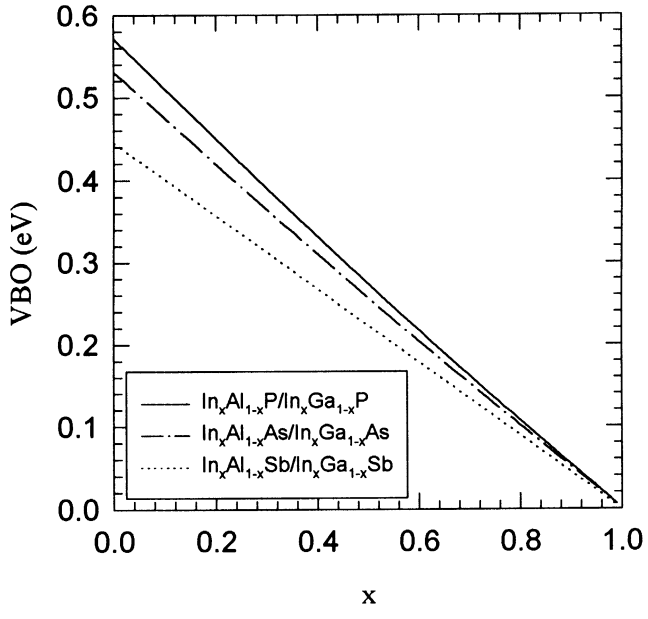

Figure 2. Variation in the valence band offsets (VBO) at $In_xAl_{1-x}M/In_xGa_{1-x}M$ (M = P, As, Sb) systems with composition.

besides $Ga_xAl_{1-x}M/Ga_yAl_{1-y}M$ and $In_xAl_{1-x}M/In_xGa_{1-x}M$ (M = P, As, Sb) systems, such as AIM/GaM (M = As_xP_{1-x}, Sb_xP_{1-x}, Sb_xAs_{1-x}), $In_xGa_{1-x}As/GaSb_xAs_{1-x}$, $In_xAl_{1-x}As/GaSb_xAs_{1-x}$, $In_xGa_{1-x}As/AlSb_xAs_{1-x}$ and $In_xAl_{1-x}As/AlSb_xAs_{1-x}$. The band offsets of these systems are listed in Table 4.

Table 4. Theoretical results of VBO (in eV) for several lattice-matched ternary alloy heterojunction systems

Heterojunction	$\Delta E_v(x)$
$AlAs_xP_{1-x}/GaAs_xP_{1-x}$	$-0.021x^2 - 0.017x + 0.569$
$AlSb_xP_{1-x}/GaSb_xP_{1-x}$	$0.053x^2 - 0.178x + 0.569$
$AlSb_xAs_{1-x}/GaSb_xAs_{1-x}$	$-0.005x^2 - 0.074x + 0.527$
$In_xGa_{1-x}As/GaSb_xAs_{1-x}$	$-0.141x^2 + 0.612x$
$In_xAl_{1-x}As/GaSb_xAs_{1-x}$	$-0.104x^2 + 0.044x + 0.529$
$In_xGa_{1-x}As/AlSb_xAs_{1-x}$	$-0.136x^2 + 0.686x - 0.529$
$In_xAl_{1-x}As/AlSb_xAs_{1-x}$	$-0.099x^2 + 0.118x$

From Table 4, one trend of the variation of the VBO at AIM/GaM (M = As_xP_{1-x}, Sb_xP_{1-x}, Sb_xAs_{1-x}) systems also can be obtained similar to Eqns (5) and (6) as follows

$$\Delta E_v^{AlSb_xAs_{1-x}/GaSb_xAs_{1-x}} < \Delta E_v^{AlSb_xP_{1-x}/GaSb_xP_{1-x}} < \Delta E_v^{AlAs_xP_{1-x}/GaAs_xP_{1-x}} \quad (7)$$

Because of the linear properties of the variation with composition in the VBO at $Ga_xAl_{1-x}M/Ga_yAl_{1-y}M$ and $In_xAl_{1-x}M/In_xGa_{1-x}M$ (M = P, As, Sb) systems and AIM′/GaM′ (M′ = As_xP_{1-x}, Sb_xP_{1-x}, Sb_xAs_{1-x}) systems, the above relationship equations (e.g. Eqns (5)–(7)) can be rewritten simply as

$$\Delta E_v^{AlSb/GaSb} < \Delta E_v^{AlAs/GaAs} < \Delta E_v^{AlP/GaP} \quad (8)$$

This equation expresses the relation of the VBO at AIM/GaM, where anion M is changed from P to Sb (the atomic number of the anion increases). For these common anion pairs, the VBO decreases as the atomic number of the anion increases. The trend in this study is in agreement with the result presented by Wei and Zunger[22] recently. Based on their p–d coupling model, the above trend can be explained as follows: due to the increase of the anion p-orbital energy and the increase of the bond lengths with an increase in anion atomic number, the p–d repulsion decreases and so also does the VBO.

Comparing our results with other experimental and theoretical data

We list our results and other theoretical and experimental results for the VBO of several lattice-matched ternary alloy heterojunctions in Table 5. It can be seen that the present results are in good agreement with experimental data.

CONCLUSIONS

In this paper, we present the theoretical results of the relative valence band position of most alloys. Some rules of average bond energy and relative valence band positions are also presented. The VBO can be obtained conveniently at the heterointerface of lattice-matched alloy semiconductors from the valence band position (i.e. the VBO parameter) data of relevant semiconductors. The calculated results show that the variation in $\Delta E_v(x)$ is nearly linear for most of the lattice-matched alloy heterojunctions and theoretical results are in very good agreement with

Table 5. Theoretical and experimental values of VBO (in eV) for several lattice-matched ternary alloy heterojunctions compared with the calculated results in this work

Heterojunction	This work	Theory	Exp.
$In_{0.52}Al_{0.48}As/In_{0.53}Ga_{0.47}As$	0.244	0.21[a], 0.23 ± 0.01[e], 0.206[h]	0.20[b], 0.22 ± 0.05[f]
$In_{0.49}Ga_{0.51}P/GaAs$	0.423	0.36[a], 0.24[h]	
$InAs_{0.95}Sb_{0.05}/GaSb$	0.450		0.67 ± 0.04[d]
$InP/In_{0.53}Ga_{0.47}As$	0.421	0.36 ± 0.05[e], 0.40[h]	0.346 ± 0.01[f], 0.43 ± 0.02[g]
$Ga_{0.7}Al_{0.3}As/GaAs$	0.159	0.132[h]	0.17 ± 0.04[j]
$In_{0.52}Al_{0.48}As/InP$	-0.177	-0.194[h]	
$Al_xGa_{1-x}As/GaAs$	$-0.002x^2 + 0.532x$		$0.55x$[j]
$InAs/In_{0.25}Ga_{0.75}Sb$	0.415	0.560[k]	
$Al_xGa_{1-x}Sb/GaSb$	$0.005x^2 + 0.439x$	$0.319x$ (heavy holes) $0.377x$ (light holes)	$(0.45 \pm 0.08)x$[l]
$Al_xGa_{1-x}Sb/AlSb$	$0.005x^2 + 0.439x - 0.443$	$0.320x - 0.320$ (heavy holes) $0.001x^2 + 0.375x - 0.376$ (light holes)	
$GaAs_{0.51}Sb_{0.49}/AlAs_{0.56}Sb_{0.44}$	0.511		0.4[m]

[a] Results from solid theory; see Ref. 9.
[b] Results from examined data using $C-V$ profiling technique; see Ref. 10.
[c] Results from examined data using $C-V$ profiling technique; see Ref. 11.
[d] Results from $C-V$ measurement; see Ref. 12.
[e] Results from empirical estimate; see Ref. 13.
[f] Results from examined data using $C-V$ profiling technique; see Ref. 14.
[g] Results from examined data using $C-V$ profiling technique; see Ref. 15.
[h] Results from empirical fit; see Ref. 16.
[i] See Ref. 17.
[j] See Ref. 18.
[k] See Ref. 19.
[l] Results from experimental data determined by a light scattering method for the low x range; see Ref. 20.
[m] See Ref. 21.

relevant experimental data. The results show that the ABE method in conjunction with the cluster expansion method is a good method for determining the VBO of alloy-type heterojunctions. The tables and figures summarizing the variation of valence band positions should be very useful in the design of novel heterostructure electronic and optical devices.

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